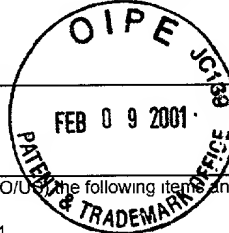


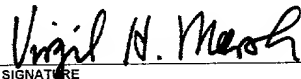
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FORM PTO-1390 U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>		LP-1799
INTERNATIONAL APPLICATION NO. PCT/EP99/05757	INTERNATIONAL FILING DATE August 9, 1999	PRIORITY DATE CLAIMED August 11, 1998
TITLE OF INVENTION  UNSATURATED OLIGOPHENOL CYANATES		
APPLICANT(S) FOR DO/EO/US  Ulrich DAUM and Alessandro FALCHETTO		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information		
<p>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. 371</p> <p>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. 371.</p> <p>3. <input type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1)</p> <p>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19<sup>th</sup> month from the earliest claimed priority date</p> <p>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))</p> <p style="margin-left: 20px;">a. <input type="checkbox"/> is submitted herewith (required only if not transmitted by the International Bureau)</p> <p style="margin-left: 20px;">b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</p> <p>6. <input type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2))</p> <p>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)).</p> <p style="margin-left: 20px;">a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau)</p> <p style="margin-left: 20px;">b. <input type="checkbox"/> have been transmitted by the International Bureau</p> <p style="margin-left: 20px;">c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired</p> <p style="margin-left: 20px;">d. <input type="checkbox"/> have not been made and will not be made.</p> <p>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3))</p> <p>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4))</p> <p>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5))</p> <p><b>Items 11 to 16 below concern other document(s) or information included:</b></p> <p>11. <input type="checkbox"/> An Information Disclosure Statement under 37 C.F.R. 1.97 and 1.98</p> <p>12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 C.F.R. 3.28 and 3.31 is included</p> <p>13. <input checked="" type="checkbox"/> A FIRST preliminary amendment <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment</p> <p>14. <input type="checkbox"/> A substitute specification.</p> <p>15. <input type="checkbox"/> A change of power of attorney and/or address letter</p> <p>16. <input type="checkbox"/> Other items or information</p>		
PCT Internationaler Vorläufiger Prüfungsbericht		
**Please enter the Preliminary Amendment before calculating the filing fee. **		



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JC05 Rec'd PCT/PTO 09 FEB 2001

U.S. Application NO. (If known, see 37 C.F.R. 1.50)		INTERNATIONAL APPLICATION NO.		ATTORNEY'S DOCKET NUMBER	
09/762579		PCT/EP99/05757		LP-1799	
17 <input checked="" type="checkbox"/> The following fees are submitted:				CALCULATIONS	PTO USE ONLY
<b>Basic National Fee (37 C.F.R. 1.492(a)(1)-(5)):</b> Search Report has been prepared by the EPO or JPO \$1,000.00  International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) \$720.00 No international preliminary examination fee paid to USPTO (37 C.F.R. 1.482) but international search fee paid to USPTO (37 C.F.R. 1.445(a)(2)) \$790.00  Neither international preliminary examination fee (37 C.F.R. 1.482) nor international search fee (37 C.F.R. 1.445(a)(2)) paid to USPTO \$1070.00  International preliminary examination fee paid to USPTO (37 C.F.R. 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$98.00				\$1,000.00	
<b>ENTER APPROPRIATE FEE AMOUNT</b>				\$1,000.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than __ 20 __ 30 months from the earliest claimed priority date (37 C.F.R. 1.492(e))				\$	
Claims	Number Filed	Number Extra	Rate		
Total Claims	-20 =		X \$18.00	\$	
Independent Claims	-3 =		X \$78.00	\$	
Multiple Dependent Claim(s) (if applicable)			X \$270.00	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$1,000.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 C.F.R. 1.9, 1.27, 1.28)				\$	
<b>SUBTOTAL =</b>				\$1,000.00	
Processing fee of \$130.00 for furnishing the English translations later than __ 20 __ 30 months from the earliest claimed priority date (37 C.F.R. 1.492(f))				\$	
<b>TOTAL NATIONAL FEE =</b>				\$1,000.00	
Fee for recording the enclosed assignment (37 C.F.R. 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 C.F.R. 3.28, 3.31) \$40.00 per property +				\$40.00	
<b>TOTAL FEES ENCLOSED =</b>				\$1,040.00	
				Amount to be refunded	\$
				Charged	\$
<p>a <input checked="" type="checkbox"/> A check in the amount of <b>\$1,040.00</b> to cover the above fees is enclosed.</p> <p>b <input type="checkbox"/> Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.</p> <p>c <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>06-1110</u>. A duplicate copy of this sheet is enclosed.</p>					
<p><b>NOTE:</b> Where an appropriate time limit under 37 C.F.R. 1.494 or 1.495 has not been met, a petition to revive (37 C.F.R. 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</p>					
SEND ALL CORRESPONDENCE TO:				 SIGNATURE	
Virgil H. Marsh Fisher, Christen & Sabol Suite 1401, 1725 K Street, NW Washington, DC 20006				Virgil H. Marsh NAME	
Tel.: 202/659-2000 February 9, 2001				23,083 REGISTRATION NUMBER	

09/762579

09/762579

JC05 Rec'd PCT/PTO 09 FEB 2001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Docket: LP-1799

Applicant : DAUM, Ulrich, et al.  
Serial No. : Not yet assigned  
[National Stage Based Upon  
PCT/EP99/05757]  
Filed : February 9, 2001  
For : UNSATURATED OLIGOPHENOL CYANATES

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Please amend this application as follows:

**IN THE SPECIFICATION:**

On page 1, between the title and the first line of the specification, insert the following:

-- This application is a continuation (national stage) application of International (PCT) Patent Application No. PCT/EP99/05757, filed on August 9, 1999, which is a continuing application of U.S. Provisional Application Serial No. 60/096,253, filed on August 12, 1998.--

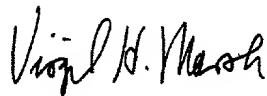
09/762579

**REMARKS**

The continuation/continuing application history of the application has been inserted on page 1 of the application.

Respectfully submitted,

Date: Jan. 9, 2001



Virgil H. Marsh  
Reg. No. 23,083

Fisher, Christen & Sabol  
Suite 1401  
1725 K Street, N.W.  
Washington, D.C. 20006  
Tel.: (202) 659-2000

FOIA b 7 - Dated: 6/25/2001

09/762579

JCO5 Rec'd PCT/PTC 09 FEB 2001

L.P. 1799, PCT

LONZA AG, Münchensteinerstrasse 38, CH-4052 Basel

Publ. No. 950000

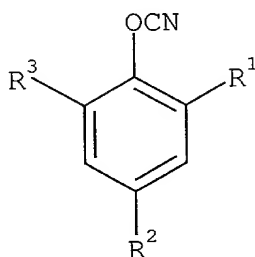
Unsaturated oligophenol cyanates

**Unsaturated oligophenol cyanates**

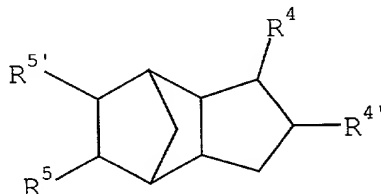
The Invention relates to oligophenol cyanates of the general formula



In this formula, A is in each case a group of the formula



and B is in each case a group of the formula



$R^1$ ,  $R^2$  and  $R^3$  on each group A independently of the others are in each case hydrogen or a bond to a group B with the proviso that each group A has either one or two bonds to B.

Both  $R^4$  and  $R^{4'}$ , and  $R^5$  and  $R^{5'}$ , on each group B independently of the others, are in each case either together a direct bond or, in any desired order, are hydrogen and a bond to a group A with the proviso that each group B has either one or two bonds to A.

The indices  $m$  and  $n$  are 0 or 1 and  $x$  is an integer from 0 to 10 with the proviso that at least one of the numbers  $m$ ,  $n$  and  $x$  is other than 0 and  $m$  and  $n$  are not both at the same time 1.

The Invention also relates in particular to mixtures of such compounds with one another and/or with those compounds of the

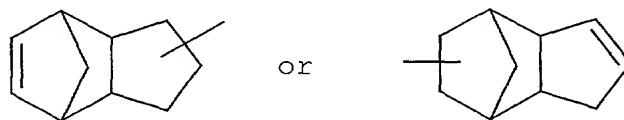
formula I in which  $m$  and  $n$  deviate from the above definitions by both being 1.

Saturated oligophenol cyanates of general formula I in which  $m$  and  $n$  deviate from the compounds of the present invention by both being 1 are known, for example, from EP-A-0 147 548 and are marketed by the Dow Chemical Co. under the designation XU71787. These compounds do not have any olefinic double bonds and are therefore able to polymerize only by cyclotrimerization of the cyanate groups or by reaction with functional groups of other compounds. The cyclotrimerization requires the presence of catalysts and/or high temperatures. In contrast it is often desirable to achieve partial curing or crosslinking by means, for example, of irradiation at room temperature. In addition, these known compounds have a relatively high viscosity, which is unfavourable for some applications.

The object of the present invention was therefore to provide oligophenol cyanates which have a low viscosity and which without further additives can be (partially) polymerized or crosslinked at room temperature by means, for example of radiation-induced free-radical reactions.

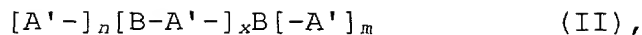
In accordance with the invention this object is achieved by the unsaturated oligophenol cyanates of the formula I in accordance with Claim 1. The molecule of these compounds has at least one olefinic double bond ( $R^4-R^{4'}$  and/or  $R^5-R^{5'}$  according to formula I) which permits free-radical addition polymerization.

The olefinic double bonds are present in groups of the formula

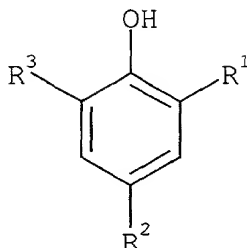


The degree of polymerization,  $x$ , lies preferably between 0 and 5 and, with particular preference, between 0 and 3.

The unsaturated oligophenol cyanates of the invention can be prepared by reacting an oligophenol of the general formula



in which A' is a group of the formula



and B, R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, R<sup>4</sup>, R<sup>4'</sup>, R<sup>5</sup>, R<sup>5'</sup>, m, n and x are as defined in Claim 1, is reacted with cyanogen chloride in the presence of a tertiary amine. Oligophenols of formula II are obtainable from Borden Chemical Inc. under the designations ESD-X1 to -X5, ESD-472C and ESD-473C. The compounds concerned here are condensation products of dicyclopentadiene (dimeric cyclopentadiene) and phenol, which are present as a mixture of isomeric and/or homologous compounds and also contain fractions of saturated compounds where m = n = 1.

The preparation of the oligophenol cyanates of the invention is preferably carried out at a temperature of less than 10 °C in a polar solvent such as butyl acetate and/or acetone or methyl ethyl ketone, for example, or in mixtures of these solvents. Particularly preferred reaction temperatures are below 0 °C - for example, -10 °C. It is advantageous to employ 1.0-1.1 mol of tertiary amine and 1.0-1.2 mol of cyanogen chloride per OH equivalent of the oligophenol II. A particularly preferred tertiary amine is triethylamine.

The unsaturated oligophenol cyanates of the invention have a low viscosity at processing temperature and produce polytriazine resins having a particularly low dielectric constant. They are particularly suitable, for example, as matrix material for the production of fibre-reinforced composites, especially for



components in aerospace technology, or as base materials for the production of printed circuit boards. Owing to their low viscosity and capacity for polymerization by means of high-energy radiation (UV, X-rays,  $\gamma$ -rays or electron beams), they are also suitable for (photo)lithographic varnishes, solder resists for circuit boards, or other radiation-curable lacquers and coatings.

The following example illustrates the preparation of the oligophenol cyanates of the invention without constituting any restriction.

### Example

Oligophenol ESD-X3 (Borden Chemical Inc.) was dissolved in n-butyl acetate/acetone (v/v = 80:20) to give a 15% strength solution. The solution was cooled to  $-10^{\circ}\text{C}$  and, at this temperature, 105% of the calculated amount of triethylamine and then, over the course of 30 minutes, 110% of the calculated amount of cyanogen chloride were added. After a further 30 minutes of reaction, the mixture was subjected to extraction with water, twice at  $30^{\circ}\text{C}$ , in order to remove the ammonium salts formed, and was then passed twice through a falling-film evaporator in order to remove the solvent and the by-product, *N,N*-diethylcyanamide.

Yield: about 100%

Properties:

viscosity: 165 mPa·s (at  $125^{\circ}\text{C}$ )

degree of reaction: (phenol  $\rightarrow$  cyanate) >98%

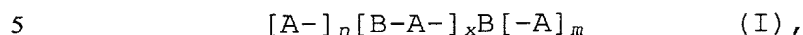
gel time: 25 min (at  $200^{\circ}\text{C}$ )

carbamates: <1%

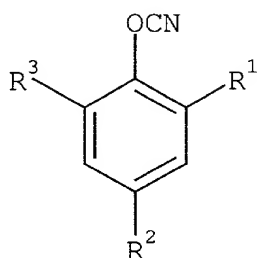
*N,N*-diethylcyanamide: <2000 ppm

# Claims

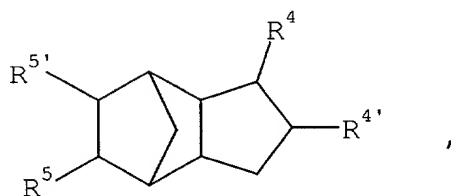
1. Unsaturated oligophenol cyanates of the formula



in which A is in each case a group of the formula



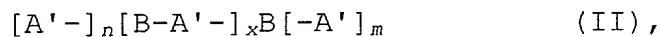
and B is in each case a group of the formula



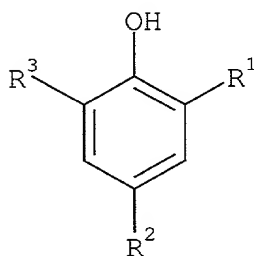
- wherein  $R^1$ ,  $R^2$  and  $R^3$  are in each case hydrogen or a bond to a group B with the proviso that each group A has either one or two bonds to B;  
 both  $R^4$  and  $R^{4'}$ , and  $R^5$  and  $R^{5'}$  are in each case either together a direct bond or are hydrogen and a bond to a group A with the proviso that each group B has either one or two bonds to A;  
 the indices  $m$  and  $n$  are 0 or 1 and  $x$  is an integer from 0 to 10 with the proviso that at least one of the numbers  $m$ ,  $n$  and  $x$  is other than 0 and  $m$  and  $n$  are not both at the same time 1, and mixtures thereof with one another and/or with those compounds of the formula I in which  $n$  and  $m$  deviate from the above definitions by both being 1.

2. Oligophenol cyanates according to Claim 1, characterized in that  $x$  is from 0 to 5.

3. Process for preparing unsaturated oligophenol cyanates according to Claim 1, characterized in that an oligophenol of the general formula



in which  $A'$  is a group of the formula



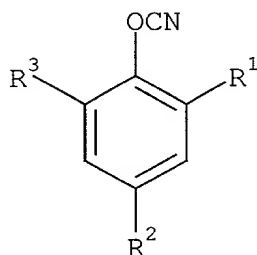
and  $B$ ,  $R^1$ ,  $R^2$ ,  $R^3$ ,  $R^4$ ,  $R^4'$ ,  $R^5$ ,  $R^5'$ ,  $m$ ,  $n$  and  $x$  are as defined in Claim 1, is reacted with cyanogen chloride in the presence of a tertiary amine.

4. Use of the unsaturated oligophenol cyanates according to Claim 1 as matrix material for fibre-reinforced composites.

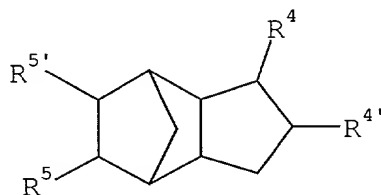
5. Use of the unsaturated oligophenol cyanates according to Claim 1 as radiation-curable varnishes, resists, lacquers and coatings.

# Abstract

The invention relates to unsaturated oligophenol cyanates of the general formula (I)  $[A-]_n[B-A-]_xB[-A]_m$  in which A is a group of formula



and B is a group of formula



where  $R^1$ ,  $R^2$  and  $R^3$  are each hydrogen or a bond with a group B, there being either one or two bonds with B; and both  $R^4$  and  $R^4'$  as well as  $R^5$  and  $R^5'$  separately or jointly represent a direct bond or hydrogen and a bond with a group A, there being either one or two bonds with A. The indices  $m$  and  $n$  are 0 or 1 but not both 1 at the same time and  $x$  is a whole number between 0 and 10, where at least one of the numbers  $m$ ,  $n$  and  $x$  is not 0.

The above unsaturated oligophenol cyanates can be prepared by reacting the corresponding oligophenols with cyanogen chloride. They have a low viscosity and owing to their double bonds are able to undergo free-radical polymerization. They are especially suitable for use as matrix materials for fibre-reinforced composites and for radiation-curable varnishes and coatings.

Type a plus sign (+) inside this box → ☐

0010/PTO Rev 6/95	U.S. Department of Commerce Patent and Trademark Office	Attorney Docket Number	LP-1799
<b>DECLARATION</b>		First Named Inventor	DAUM
		<b>COMPLETE IF KNOWN</b>	
		Application Number	
		Filing Date	
		Group Art Unit	
<input checked="" type="checkbox"/> Declaration Submitted with Initial Filing      OR <input type="checkbox"/> Declaration Submitted After Initial Filing		Examiner Name	
		As a below named inventor, I hereby declare that:	
My residence, post office address, and citizenship are as stated below next to my name			
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled.			
<div>UNSATURATED OLIGOPHENOL CYANATES</div>			
(Title of the invention)			
the specification of which			
<input checked="" type="checkbox"/> is attached hereto			
OR			
<input type="checkbox"/> was filed on (MM/DD/YYYY) <input type="text"/> as United States Application Number or PCT International Application			
Number <input type="text"/> and was amended on (MM/DD/YYYY) <input type="text"/> (if applicable)			
I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.			
I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § .56.			
I hereby claim foreign priority benefits under Title 35, United States Code §119 (a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed			
Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed Copy Attached? YES NO
PCT/EP99/09477	International	08/09/1999	<input type="checkbox"/> <input checked="" type="checkbox"/> <input type="checkbox"/>
98202692.4	Europe	08/11/1998	<input type="checkbox"/> <input checked="" type="checkbox"/> <input type="checkbox"/>
60/096,523	U.S. Provisional	08/12/1998	<input checked="" type="checkbox"/> <input type="checkbox"/> <input checked="" type="checkbox"/>
			<input type="checkbox"/> <input type="checkbox"/> <input type="checkbox"/>
<input type="checkbox"/> Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:			
I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.			
Application Number(s)	Filing Date (MM/DD/YYYY)	<input type="checkbox"/> Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.	

## DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations 1.56 which became available between the filing date of the prior application and the national or PCT filing date of this application

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
60/096,253	PCT/EP99/05757	08/09/1999 08/12/1998	
<input type="checkbox"/> Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto			

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith

Firm Name	<b>Fisher, Christen &amp; Sabol</b>	Payor Number (if applicable)	
Name	Registration Number	Name	Registration Number
Virgil H. Marsh	23,083		
Kara M. Armstrong	38,234		
<input type="checkbox"/> Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto			

<input checked="" type="checkbox"/> Please direct all correspondence to	Name	Virgil H. Marsh	
Address <b>Fisher, Christen &amp; Sabol</b>			
Address <b>Suite 1401, 1725 K Street, N.W.</b>			
City	Washington	State	D.C.
Country	USA	Telephone	(202)659-2000
		Fax	(202)659-2015
I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon			

Name of Sole or First Inventor:				<input type="checkbox"/> A petition has been filed for this unsigned inventor			
Given Name	Ulrich	Middle Initial		Family Name	DAUM	Suffix	
Inventor's Signature	<i>Ulrich</i>			Date			01/26/01
Residence: City	Hofstetten	Country	Switzerland	Citizenship	German		
Post Office Address: Chöpliweg 17							
City	Hofstetten	Postal Code	CH-4114	Country	Switzerland	Applicant Authority	
<input checked="" type="checkbox"/> Additional inventors are being named on supplemental sheet(s) attached hereto							

